

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant: Hillion et al.  
Serial No.: 10/802, 042  
Filed: March 17, 2004

Examiner: Price, Elvis O.  
Group Art Unit: 1621

For : PROCESS FOR THE ALCOHOLYSIS OF ACID OILS OF VEGETABLE OR ANIMAL  
ORIGIN.

**DECLARATION UNDER 37 C.F.R. §1.132**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir :

I, Pierre BECCAT, being duly warned, declare and say as follows:

THAT, I am a french citizen holding the titles of Engineer delivered by "Ecole Nationale Supérieure des Ingénieurs Electriciens de Grenoble" in 1987, of "Docteur ès Sciences" delivered "Université de Grenoble" in 1990, residing at 69360 Communay, France, 7 allée des Merisiers.

THAT, I have been engaged on research by "Institut Français du Pétrole" in their "Physics and Analysis" Division from 1991 to 2006 and in their "Catalysis and Separation" Division since 2006.

THAT, I have been continuously and actively in charge of researches in the fields of refining and petrochemicals. I am since May 2006 the manager of the Division "Catalysis and Separation".

I declare further :

THAT, I am familiar with the content of U.S. Patent Application Serial No 10/802, 042.

THAT, I know the teaching of the patent which number is US 6,245,727.

THAT in order to support patentability of the present application, I have supervised the following experiments :

Example : Transesterification of vegetable oil by methanol in a batch reactor

The transesterification reaction of vegetable oil by methanol was performed in a stirred batch reactor. Before each test, to ensure that the reactor wall do not interfere with the catalyst being evaluated, the reactor wall were passivated by a 2g/L NaOH solution for 2 hours, then rinsed with water and finally filled with distilled water, closed and heated up overnight at 200 °C. A blank test was done (test 0) , in the absence of any catalyst, to be sure of the efficiency of the passivation procedure.

For each further test, a mixture 50/50 wt% of methanol and vegetable oil were introduced into the reactor together with the catalyst.

In test 1 and test 2, the catalyst was a strongly acidic ion exchanger resin, Amberlite XE 356, the same as in example 1 in Gutsche *et al.* (US 6.245.727 B1), hereafter referred to as Catalyst A

In test 3, the catalyst was a ZnO, Al<sub>2</sub>O<sub>3</sub>, ZnAl<sub>2</sub> mixture according to the present application, hereafter referred to Catalyst B

The reactor was closed and heated up to the desired temperature, stirring at 800 rpm. The starting point of the reaction was taken once this target temperature was reached. Samples were then taken at t = 0, 2, 4 and 6 hours and analysed by gel permeation chromatography in order to determine the relative amounts of methyl esters, mono-, di- and triglycerides.

To describe the test results, the following acronyms and terms are used, as defined below :

- \* TG : triglycerides remaining in the reactor effluent, analysed through GPC technique.
- \* DG : diglycerides remaining in the reactor effluent, analysed through GPC technique.
- \* MG : monoglycerides remaining in the reactor effluent, analysed through GPC technique.
- \* FAME : Fatty acids methyl esters remaining in the reactor effluent, analysed through GPC technique.

**Test results**

The results of the different tests are given in Table 1.

Table 1 : Composition of the reaction medium at t = 6 hours

	catalyst	temperature	water content (ppm)	TG	DG + esterified sterols	MG	FAME + sterols
Blank	-	180	900	86,7	9,0	0,4	3,9
Test 1	30 g of Catalyst A	80	590	93,5	3,0	0,4	3,0
Test 2	30 g of Catalyst A	120	560	70,0	10,8	1,4	17,7
Test 3	5 g of Catalyst B	190	940	21,9	19,1	10,9	48,0

These results show that the Catalyst A has a much weaker activity for transesterification reaction than the Catalyst B catalyst of the present invention and is therefore not suitable for this application.

Example 1 in US 6,245,727 shows 98,3% conversion reached after 5 hours, at 130°C. Our tests show that after 6 hours at 120 °C, FAME, which is the main reaction product, is only 17,7 %, and only 30 % of the triglycerides have been converted.

Moreover, it is well known by one skilled in the art that this kind of catalyst (ion exchanger resin) can not be used at higher temperatures since the resin decomposes and is not stable at these temperatures.

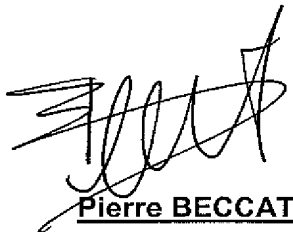
For the catalyst used in the present application, with 6 times less quantity, at 190 °C which is within the temperature range of the IFP patent, 80 % of triglycerides have been converted. Moreover, the test 3 gives better results, even with a higher water content that acts as a catalyst inhibitor.

Thus these comparative examples show a better conversion rate with the catalyst of the present invention.

The undersigned declares further that all statements made herein of this own knowledge are true and that all statements made on information or belief are believed to be true; and further

that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 Title 18 of United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Solaize, October 1<sup>st</sup>, 2007.



Pierre BECCAT